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Spectroscopic study of formation and dissociation processes of methane hydrate in porous media

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Natural gas-hydrates, whose guest gas is primarily of methane, are widely distributed in oceanic and permafrost sediments. These gasses in natural gas-hydrates are expected to become a new energy resource. Therefore, gas production methods for natural gas from hydrate-bearing sediments are currently being developed. In gas production process by dissociating natural gas hydrates from sediments, initial/absolute permeability of water/gas in sediments is very important. To understand manner of hydrate growth in pore spaces of sediment is very important for mechanism of hydrate formation, but also for dissociation of hydrate in gas production form natural-gas hydrate sediments. We observed processes of methane (CH4) hydrate formation and dissociation in unconsolidated sandy media by using an all attenuated total reflection infrared (ATR-IR) spectroscopy. IR absorption region of H_2O molecules is very useful to evaluate hydrogen-bonded (HBd) states of O-H group (OH). With increasing HBd OH species (namely, phase transition from liquid to solid), ATR-IR absorption band of OH shifts to lower-wavenumber region. On the other hands, the OH band shifts to higher-wavenumber region during decreasing HBd OH species (phase transition from solid to liquid phase). From ATR-IR spectra during CH4 hydrate formation, two discrete growths were observed after pressurization. This two-step growth was not reported in bulk hydrate formation. At the first step, randomly hydrate double-exponentially increased after nucleation. The second discrete growing step randomly occurs. Observations of dissociation process of CH4 hydrate were performed by depressurizing method. In dissociation process, discontinuous changes of OH band were observed by rapid-depressurizing. The discontinuous change is considered to show ice formation in the porous media. In our experiments, we found not only thermal, but also spectroscopic evidences of ice formation in pores during hydrate dissociation. Moreover, due to assess of center of gravity of the OH band, it was found that ATR-IR spectra of hydrate provide us with the quantitative information of the hydrate dissociation behavior in porous media.